

# Dynamic nonlinear (cubic) susceptibility in quantum Ising spin glass

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## Abstract

Dynamic nonlinear (cubic) susceptibility in quantum d-dimensional Ising spin glass with short-range interactions is investigated on the basis of quantum droplet model and quantum-mechanical nonlinear response theory. Nonlinear response depends on the tunneling rate for a droplet which regulates the strength of quantum fluctuations. It shows a strong dependence on the distribution of droplet free energies and on the droplet length scale average. Comparison with recent experiments on quantum spin glasses like disordered dipolar quantum Ising magnet is discussed .

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The dynamics of glassy systems is an attractive and rapidly developing field of physics [1-5]. Spin glasses and quantum spin glasses are very interesting systems for theoretical and experimental investigation of dynamic phenomena [2-4]. Two major theoretical description of spin glass phenomena have evolved over the past twenty years: the mean field theory based on the Parisi replica symmetry-breaking approach and the droplet model based on renormalization group arguments [1-6]. The two pictures give very different physical interpretation of observable spin-glass phenomena. In this paper we investigate theoretically nonlinear cubic dynamic susceptibility as function of frequency and temperature in the Ising spin glass in a transverse field in terms of quantum droplet model at very low temperatures (quantum regime). The quantum phase transition is governed by quantum fluctuations of the system which may tunnel from one local minimum of the free energy to another; new physical effects such as quantum channel of relaxation appear. There are few theoretical studies on the nonlinear static response in quantum spin glasses [7-10] and almost no studies on the dynamic nonlinear response [3]. In [3] dynamic nonlinear response of a quantum spin glass was found frequency independent and nonsingular in quantum critical regime in contrast to the behavior in usual spin glass. There are experimental data on the nonlinear dynamic response in classical [11-13] and quantum [14] spin glasses investigated by Fourier-transform technique. The third-order nonlinear susceptibility is negative and diverges at an ordinary spin glass transition temperature  $T_g$  from both the upper and the lower sides. But when  $\chi'_3$  is measured by a finite probing frequency the response falls out equilibrium before the transition temperature and does not diverge at  $T_g$ . Then  $\chi'_3(\omega)$  shows a maximum at  $T \simeq T_f(\omega)$  where  $T_f(\omega)$  is the freezing temperature which is the upper bound on  $T_g$  and  $T_g = T_f(\omega \rightarrow 0)$ . Such behavior was observed, for example, for classical Ising spin glass  $\text{Fe}_{0.5}\text{Mr}_{0.5}\text{TiO}_3$  [13]. W. Wu et al. [14] measured nonlinear susceptibility  $\chi'_3(\omega, T)$  in quantum spin glass (the diluted dipolar-coupled Ising spin glass  $\text{LiHo}_{0.167}\text{Y}_{0.833}\text{F}_4$  in the transverse field) tuning transverse field  $\Gamma$  from the  $\Gamma = 0$  classical to the  $T = 0$  quantum limit. At  $mK$  temperatures they found a clear dynamic signature of the spin glass to paramagnet transition whether dominated by thermal or quantum fluctuations. In [14] it was shown that the  $\chi'_3$  depends on frequency for  $\omega > 10Hz$ . However it depends very weakly on  $\omega$  for  $\omega < 10Hz$ . There is a crossover between high  $\omega$  ( $\omega$  - dependent) and low  $\omega$  ( $\omega$  - independent) behaviors. Nonlinear susceptibility contains a diverging component which dominates at  $T = 98mK$ , but disappears by  $25mK$ . The  $\chi'_3(\omega)$  does not diverge but shows a maximum at  $T_f(\omega)$ .  $\chi'_3(\omega)$  measured at a higher temperature and lower transverse field has a larger maximum than  $\chi'_3(\omega)$  measured at a lower temperature and larger transverse field. The analysis of these experimental data seems not clear

[15] because frequencies used in the experiments [14] are not sufficiently low such as to determine the equilibrium behavior of system. Contrary to the theoretical expectations, quantum transitions may be qualitatively different from thermally driven transitions in real spin glasses. Recently the linear dynamic susceptibility (in-phase and out of phase components) at  $T = 0$  was investigated theoretically for the Ising spin glass in a transverse field in terms of quantum droplet model by M. J. Thill and D. A. Huse [6]. A basic assumption of the droplet picture is that the spin glass dynamics is governed by large-scale excitations whose relaxation time increases with length scale. In previous papers [16] we have calculated the real and imaginary parts of linear dynamic susceptibility in the same model, as in [6], for very low nonzero temperatures using the general linear response theory of magnetic dispersion and absorption phenomena for quantum systems by Kubo and Tomita [17]. We note that in [6] the real part of the cubic nonlinear ac susceptibility was defined as the in-phase  $3\omega$  magnetization response  $M(3\omega)$  to a small time-dependent applied field  $h \cos(\omega t)$

$$\chi'_3 = \lim_{h \rightarrow 0} \frac{24M(3\omega)}{h^3 V} \quad (1)$$

where  $V$  is the sample volume. The authors of [6] gave some expression for  $\chi'_3$  they only expect at zero temperature.

The full nonlinear response theory despite its generality and importance is of limited practical value because it is mathematically difficult. It is necessary to make approximations, as the well-known perturbation expansion of the time-evolution operator, using Hamiltonian with some small parameter. Nonlinear response theory was developed and described, for example, in [17-22]. Here we summarize briefly the theory of higher-order dynamic response. It is based on the Hamiltonian

$$\hat{\mathcal{H}}_t = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_1 = \hat{\mathcal{H}}_0 - \hat{A}_j F_j(t), \quad t \geq t_0 \quad (2)$$

where  $\hat{\mathcal{H}}_0$  is nonperturbation Hamiltonian of system,  $\hat{\mathcal{H}}_1$  is perturbation Hamiltonian which describes the interaction between the Heisenberg operators  $\hat{A}_j$  (material operators) and the external perturbation  $\hat{F}_j$ . At time  $t > t_0$  one is interested in the expectation value of the Heisenberg operator  $\hat{B}_i$  which is given by

$$\langle \hat{B}_i(t) \rangle = \text{Tr}[\rho_0 \hat{B}_i(t)] = \text{Tr}[\hat{\rho}(t) \hat{B}_i] \quad (3)$$

where the density matrix  $\hat{\rho}(t) = \hat{U}(t, t_0) \hat{\rho}(t_0) \hat{U}^\dagger(t, t_0)$ . The time-evolution operator  $\hat{U}$  satisfies the Schrödinger equation  $i\hbar \frac{d\hat{U}}{dt} = \hat{\mathcal{H}} \hat{U}$ . It is difficult to find an expression for  $\hat{U}$  in closed form. It was used that  $\hat{\mathcal{H}}_1(t)$  is in some sense small. We define the total response of the system at time  $t$  to the external force  $F_j$  as the difference

$$\Delta B_i(t) \equiv \langle \hat{B}_i(t) \rangle - \langle \hat{B}_i \rangle_0 \quad (4)$$

where the subscript zero on expectation values refers to the equilibrium expectations. One can then understand the behavior of the system in terms of the dynamical response. Using aforementioned expressions [2-3] the dynamical response can be written through the third order in the perturbation  $\hat{\mathcal{H}}_1$  in the following form [20]

$$\begin{aligned} \langle \hat{B}_i(t) \rangle - \langle \hat{B}_i \rangle_0 = \Delta B_i(t) \simeq & \int_{t_0}^t dt' \varphi_{ij}(t-t') F_j(t') + \\ & \int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \varphi_{ijk}(t-t_2, t_1-t_2) F_k(t_1) F_j(t_2) + \\ & \int_{t_0}^t dt_1 \int_{t_0}^{t_1} dt_2 \int_{t_0}^{t_2} dt_3 \varphi_{ijkl}(t-t_3, t_2-t_3, t_1-t_2) F_e(t_1) F_k(t_2) F_j(t_3) + \dots \end{aligned} \quad (5)$$

where  $\varphi_{ij}$ ,  $\varphi_{ijk}$ ,  $\varphi_{ijkl}$  are the first-, second- and third order response functions,

$$\varphi_{ij}(t-t') = \frac{1}{i\hbar} \langle [\hat{A}_j, \hat{B}_i(t-t')] \rangle_0, \quad (6)$$

$$\varphi_{ijk}(t-t_2, t_1-t_2) = \frac{1}{(i\hbar)^2} \langle [\hat{A}_j, [\hat{A}_k(t_1-t_2), \hat{B}_i(t-t_2)]] \rangle_0, \quad (7)$$

$$\varphi_{ijkl}(t-t_3, t_2-t_3, t_1-t_2) = \frac{1}{(i\hbar)^3} \langle [\hat{A}_j, [\hat{A}_k(t_1-t_2), [\hat{A}_l(t_2-t_3), \hat{B}_i(t-t_3)]]] \rangle_0. \quad (8)$$

Here we have employed the summation convention over repeated indices and cyclic invariance of the trace. The expressions (6-8) can be written in a more revealing form if we set  $t_0 = -\infty$  and change integration variables including an adiabatic switching factor if necessary. We take (instead of  $t, t_1, t_2, t_3$ )  $\tau_1 = t-t_1, \tau_2 = t_1-t_2$  time differences further. Ordinary linear response theory utilized only  $\varphi_{ij}(\tau_1)$  and it is the simplest approximation to the full theory of linear dynamic response [23]. Using new variables we may write the expressions for response functions in the form

$$\varphi_{ij}^{(1)} = -\frac{1}{i\hbar} \langle [A(\tau), B(0)] \rangle, \quad (9)$$

$$\varphi_{ijk}^{(2)} = \frac{1}{(i\hbar)^2} \langle [[A(\tau_1 + \tau_2), A(\tau_2)], B] \rangle, \quad (10)$$

$$\varphi_{ijkl}^{(3)} = -\frac{1}{(i\hbar)^3} \langle [[[A(\tau_1 + \tau_2 + \tau_3), A(\tau_2 + \tau_3)], A(\tau_3)], B] \rangle \quad (11)$$

where the bracket  $\langle \dots \rangle$  denotes an expectation value with respect to the equilibrium ensemble. The useful interpretation is generated from eq. (5) in the case that  $t_0 = -\infty$  if we consider the external force  $F$  is constant and vanishes for  $t \geq 0$ . For  $t = 0$  the system is in partial equilibrium and starts to relax to equilibrium. It is convenient to write nonlinear response for this case (initial value case [21]) as

$$\langle \hat{B}(t) \rangle - \langle \hat{B} \rangle_0 = R^{(1)}(t)F + \frac{1}{2}R^{(2)}(t)FF + \frac{1}{3}R^{(3)}(t)FFF + \dots \quad (12)$$

where  $R^\alpha(t)$  are the relaxation functions,

$$R^{(1)}(t) = \int_0^\infty d\tau \varphi_{ij}^{(1)}(\tau_1), \quad (13)$$

$$R^{(2)}(t) = \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \varphi_{ijk}^{(2)}(\tau_2, \tau_1 + \tau_2), \quad (14)$$

$$R^{(3)}(t) = \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3 \varphi_{ijkl}^{(3)}(\tau_3, \tau_2 + \tau_3, \tau_1 + \tau_2 + \tau_3). \quad (15)$$

In this form response may describe relaxation of the system. If response function  $\varphi(t)^{(1)}$  vanishes as  $t \rightarrow \infty$ , then  $\varphi(t)^{(1)} = -\frac{\delta}{\delta t}R^{(1)}(t)$ , so  $R^{(1)}(t)$  contains much more information than the response function.

Let the *ac* magnetic field  $h_\omega = h \cos(\omega t)$  be applied to a magnetic system. The magnetization nonlinear response  $M(\omega, t) = \sum_{k=1}^n \{\Theta'_k \cos(k\omega t) + \Theta''_k \sin(k\omega t)\}$  to harmonic magnetic field  $h$  contains only odd harmonic;  $\Theta'_1 \sim \chi'_1 h$ ,  $\Theta'_3 \sim \chi'_3 h^3$ , etc [11]. In expression for  $M(\omega, t)$  the magnitudes  $\Theta'_k$  and  $\Theta''_k$  are real and imaginary parts of harmonic amplitudes respectively. For the general theory of nonlinear processes one can evaluate [11]

$$\begin{aligned} \Theta'_1 &= \chi'_1(\omega t)h + [\chi'_3(\omega, 0, \omega)]\frac{h^3}{4} + [4\chi'_5(\omega, 0, \omega, 0, \omega) + 2\chi'_5(\omega, 0, \omega, 2\omega, \omega) + \\ &2\chi'_5(\omega, 2\omega, \omega, 0, \omega) + \chi'_5(\omega, 2\omega, \omega, 2\omega, \omega) + \chi'_5(\omega, 2\omega, 3\omega, 2\omega, \omega)]\frac{h^5}{16} + \dots \end{aligned} \quad (16)$$

$$\begin{aligned} \Theta'_3 &= \chi'_3(3\omega, 2\omega, \omega)\frac{h^3}{4} + [\chi'_5(3\omega, 2\omega, 3\omega, 2\omega, \omega) + \chi'_5(3\omega, 4\omega, 3\omega, 2\omega, \omega) \\ &2\chi'_5(3\omega, 2\omega, \omega, 0, \omega) + \chi'_5(3\omega, 2\omega, \omega, 2\omega, \omega)]\frac{h^5}{16} + \dots, \end{aligned} \quad (17)$$

$$\Theta'_5 = \chi'_5(5\omega, 4\omega, 3\omega, 2\omega, \omega)\frac{h^5}{16} + \dots \quad (18)$$

The measurement of all the harmonic amplitudes  $\Theta_k$  gives a measurement of the susceptibilities  $\chi_a$  in two limits: a) if  $\chi'_1 h \gg \chi'_3 h^3 \gg \chi'_5 h^5$  the back reaction is negligible and each harmonic measures the susceptibility of the same order; b) in the static ( $\omega \rightarrow 0$ ) limit the solution of the linear system [15] fully accounts for the back reaction. In the absence of *ac* magnetic field, the back reaction can be made small so that the dynamic susceptibilities can be obtained from eq. (15). In a more compact notation we may write

$$M(\omega, t) \sim [M_0 + M_\omega + M_{3\omega} + \dots] + (\text{complex conjugate}) \quad (19)$$

where  $M_0$  is the equilibrium magnetization in zero field;  $M_\omega$  is the  $\omega$ -magnetization response;  $M_{3\omega}$  is the  $3\omega$ -magnetization response and so on.

The expressions (9-11) may be considered as solution of the corresponding quantum equations considered above. The external *ac* field we assume classical value. This field interacts with quantum system and system behavior is determined by quantum laws. We shall focus on the real part of the third-order nonlinear dynamic susceptibility  $\chi'_3(3\omega, 2\omega, \omega)$  and denote it as  $\chi'_3(\omega)$ . In this paper we are interested in the response when the *ac* magnetic field is applied in *z*-direction;  $\chi_1 = \chi_{zz}$  and so on. In formulas (2-11) both  $A_i$  and  $B_i$  are the magnetic dipole moment operators. Considering the initial value case [21] we suppose, like in [6], that the system is in equilibrium with a small time-independent field  $h = h_z$  for  $t \leq 0$  and that the external field is turned off at  $t = 0$ , then for  $t \geq 0$  the induced magnetization of the sample in *z*-direction to first order of perturbation theory is given by  $M(t) - M_0 \approx R^{(1)}(t)h$ , where a relaxation function

$$R^{(1)}(t) = \int_0^\infty \varphi^{(1)}(\tau) d\tau \quad (20)$$

and the first order response function is [20]

$$\varphi_{ij}^{(1)}(\tau) \approx -\frac{1}{i\hbar} \langle [M_i(\tau), M_j] \rangle. \quad (21)$$

The higher-order response functions are given by

$$\varphi_{ijk}^{(2)}(\tau_1, \tau_2) \approx \frac{1}{(i\hbar)^2} \langle [[M_i(\tau_1 + \tau_2), M_j(\tau_2)], M_k] \rangle; \quad (22)$$

$$\varphi_{ijkl}^{(3)}(\tau_1, \tau_2, \tau_3) \approx -\frac{1}{(i\hbar)^3} \langle [[[M_i(\tau_1 + \tau_2 + \tau_3), M_j(\tau_2 + \tau_3)], M_k(\tau_3)], M_l] \rangle. \quad (23)$$

The linear and the nonlinear dynamic susceptibilities (admittances in the spectral representation [22]) may be found through response functions (21-23). In order to find the complete expression for susceptibility, we should use its symmetry and causality properties [22].

The nonlinear susceptibilities may be chosen so that these susceptibilities were symmetrical relative to simultaneous permutation of tensor indices and corresponding to them arguments, for example, the second rank tensors are  $\chi_{ijk}(\omega_1, \omega_2) = \chi_{ikj}(\omega_2, \omega_1)$ , and the fourth rank tensors are

$$\chi_{ijkl}(\omega_1, \omega_2, \omega_3) = \chi_{ikjl}(\omega_2, \omega_1, \omega_3) = \chi_{ijlk}(\omega_1, \omega_3, \omega_2) = \dots \quad (24)$$

according to causality property

$$\chi_{ij} = 0 \text{ for } \tau_1 < \max(\tau_2, \tau_3, \dots). \quad (25)$$

Linear and nonlinear dynamic susceptibilities are given by

$$\chi_{ij}(\omega) = \int_0^\infty d\tau \varphi_{ij}^{(1)}(\tau) e^{i\omega\tau} \quad (26)$$

$$\chi_{ijk}(\omega_1, \omega_2) = \frac{1}{2!} \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \left\{ \varphi_{ijk}^{(2)}(\tau_1, \tau_2) e^{i(\omega_1 + \omega_2)\tau_1 + i\omega_1\tau_1} \right\} \quad (27)$$

$$\begin{aligned} \chi_{ijkl}(\omega_1, \omega_2, \omega_3) &= \frac{P_3}{3!} \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3 \varphi_{ijkl}^{(3)}(\tau_1, \tau_2, \tau_3) \times \\ &\times \exp[i(\omega_1 + \omega_2 + \omega_3)\tau_1 + i\omega_2\tau_2 + i\omega_3\tau_3] \end{aligned} \quad (28)$$

In eq. (28)  $P_3$  means the summation over all permutations of the subscripts  $(\omega_1 j)$ ,  $(\omega_2 k)$  and  $(\omega_3 l)$  [22]; response functions  $\varphi$  are given by expressions (21-23).

In particular, the second harmonics generation process is characterized by tensor  $\chi_{ijk}(\omega, \omega)$ . If  $\omega_1 = \omega_2 = \omega_3 = \omega$ , the triple frequency  $3\omega$  is formed; the frequency tripling is described by tensor  $\chi_{ijkl}(\omega, \omega, \omega)$  (we note it as  $\chi'_3(\omega)$ ).

The droplet model of classical Ising spin glass was considered by D. S. Fisher and D. A. Huse [4]. The features of this model are described also, for example, in [23]. In the droplet model there are only two pure thermodynamical states related to each other by a global spin flip. In magnetic field there is no phase transition. A droplet is an excited cluster in an ordered state where all the spins are inverted. The natural scaling ansatz for droplet free energy  $\epsilon_L$  (which are considered to be independent random variables) is  $\epsilon_L \sim L^\theta$ ,  $L \geq \zeta(T)$ ;  $\zeta$  is the correlation length,  $L$  is the length scale of droplet and  $\theta$  is the zero temperature thermal exponent,  $\theta \leq (d-1)/2$ . One droplet consists of order  $L^d$  spins. Below the lower critical dimension  $d_l$ ,  $\theta < 0$ ; above  $d_l$  one has  $\theta > 0$ .

Recently M. J. Thill and D. A. Huse [6] have shown that the  $d$ -dimensional quantum ising spin glass in a transverse field with Hamiltonian

$$\mathcal{H} = - \sum_{i,j} \mathcal{I}_{ij} S_i^z S_j^z - \Gamma \sum_i S_i^x \quad (29)$$

(where  $S_i$  are the Pauli matrices,  $\Gamma$  is the strength of the transverse field and the nearest neighbor interactions  $\mathcal{I}_{ij}$  are independent random variables of mean zero) can be represented as the Hamiltonian of the independent quantum two-level systems (low energy droplets) of the form

$$\mathcal{H} = \frac{1}{2} \sum_L^{\sim} \sum_{D_L} \left( \epsilon_{D_L} S_{D_L}^z + \Gamma_L S_{D_L}^x \right) \quad (30)$$

where  $S_{D_L}^z$  and  $S_{D_L}^x$  are the Pauli matrices representing the two states of the droplet; the sum is over all droplets  $D_L$  at length scale  $L$  and over all length scales  $L$ , and

$$\sum_L^{\sim} \sim \int_{L_0}^\infty \frac{dL}{L} \quad (31)$$

with a short-distance cutoff  $L_0$ . The value  $\Gamma_L$  regulates the strength of quantum fluctuations ( $\Gamma_L \rightarrow 0$  corresponds to the classical limit).

$$\Gamma_L = \Gamma_0 e^{-\sigma L^d} \quad (32)$$

is the tunneling rate for a droplet of linear size  $L$ ,  $\Gamma_0$  is the microscopic tunneling rate;  $\sigma$  is defined from the equation  $2K = \sigma L^d$  where  $2K$  is the surface free energy of an interface between the two droplet states, so  $\sigma$  is a reduced surface tension for this interface;  $\sigma$  is approximately the same for all droplet. We will assume  $\Gamma_L$  is the same for all droplets of scale  $L$ . The droplet excitations have a broad distribution of their free energies at scale  $L$  for large  $L$  in a scaling form [4,6]

$$P_L(\epsilon_L)d\epsilon_L = \frac{d\epsilon_L}{\gamma(T)L^\theta} \mathcal{P}\left(\frac{\epsilon_L}{\gamma(T)L^\theta}\right), L \rightarrow \infty. \quad (33)$$

It is assumed that  $P_L(x \rightarrow 0) > 0$ ,  $P_L(0) - P_L(x) \sim x^\phi$  at  $x \rightarrow 0$ .  $\gamma(T)$  is a generalized temperature dependent stiffness modulus which is of order of characteristic exchange  $\mathcal{I} = \overline{(\mathcal{I}_{ij}^2)}^{\frac{1}{2}}$  at  $T = 0$  and vanishes for  $T \geq T_g$ .

There is a crossover length scale,  $L^*(T)$ , defined by condition  $\Gamma_{L^*(T)} = k_B T$  or  $L^*(T) = \left(\frac{1}{\sigma} \log \frac{\Gamma_0}{k_B T}\right)^{1/d}$ . For droplets with  $L \ll L^*(T)$  and  $\Gamma_L \gg k_B T$  the energy  $\sqrt{\epsilon_L^2 + \Gamma_L^2}$  is always more than  $k_B T$  and thermal fluctuations are insignificant at temperature  $T$ . Droplets with  $L \gg L^*(T)$  have  $\Gamma_L \ll k_B T$  and behave classically. The large droplets ( $\epsilon_L \leq k_B T$ ,  $\Gamma_L \leq k_B T$ ) are thermally active. At low  $T$  only a small fraction of droplets is thermally active, but many low- $T$  static properties are dominated by these droplets at the crossover length  $L^*(T)$ .

The total magnet moment of a droplet will scale as  $qL^{\frac{d}{2}}$  where  $q$  a random number with mean zero and  $\overline{q^2} \sim q_{EA}$ ,  $q_{EA} = \overline{(S_i^z)^2}$  is the Edwards-Anderson order parameter [4]. The total magnetization  $M$  of the sample will be

$$M = \sum_L \frac{\tilde{V}}{L^d} \sum_{D_L} \langle S_{D_L}^z \rangle q L^{\frac{d}{2}} \quad (34)$$

and

$$m = \frac{M}{V} = \sum_L \overline{\langle S_{D_L}^z \rangle q L^{\frac{-d}{2}}} \quad (35)$$

where  $\overline{\langle S_{D_L}^z \rangle q L^{\frac{-d}{2}}}$  means the average over the droplets energies  $\epsilon_L$ . The static susceptibilities are defined in terms of the expansion of magnetization into Taylor series approximately as

$$M = \chi_1 h - \chi_3 h^3 + \dots \quad (36)$$

For enough small external field it is possible to be restricted by few terms of this expansion. The susceptibilities are got as derivatives with respect to  $h$  at  $h = 0$  :  $\chi_1 = \frac{\partial m}{\partial h} \Big|_{h=0}$ ,  $\chi_3 = \frac{\partial^3 m}{\partial h^3} \Big|_{h=0}$ .

M. J. Thill and D. A. Huse have calculated static linear and nonlinear susceptibilities for droplet system described by the Hamiltonian (29). The static linear susceptibility diverges at  $T = 0$  below the lower critical dimension  $d_l$ . The static



nonlinear susceptibility diverges in all dimensions  $d$ . The static linear susceptibility appears to start away from the nonzero constant  $T = 0$  value decreasingly versus  $T$  to lowest order [6].

Now using the quantum droplet model of the short-range Ising spin glass in a transverse field and quantum-mechanical case  $\beta\Gamma_L \gg 1$  (quantum regime) we calculate the third order nonlinear dynamic response at very low finite temperatures.

When we consider the droplets at finite temperatures they may have two characteristic rates, (a) the Rabi frequency (is of order  $\Gamma_L$ ) and (b) the rate of classical activation over energy barrier  $B$  for annihilation and creation of the droplet excitations [6]

$$t \sim \tau_0 e^{\frac{B}{k_B T}}, \quad (37)$$

where  $B \sim \Delta L^\psi$ ,  $0 \leq \psi \leq d-1$ ,  $\psi$  is some exponent [4],  $\Delta$  is a barrier energy at  $T \ll T_g$ ,  $\Delta \sim \mathcal{I}$ ;  $\tau_0$  is a microscopic time. There is a complicated dynamical classical-to-quantum crossover depending on temperature, frequency of  $ac$  external field and length scale  $L$ . According to [6] the crossover dynamic length is determined from the condition  $\Gamma_L^{-1} = T$ , i.e.

$$L_{dyn}^*(T) \sim \left( \frac{\sigma}{\Delta} k_B T \right)^{\frac{1}{\psi-d}} \quad (38)$$

The system behaves presumably classically or quantum mechanically when the dominant length scale  $L$  is above or below  $L_{dyn}^*$  for frequency  $\omega$ .

Now we consider dynamic third-order susceptibility  $\chi'_3(\omega, T)$  at finite very low temperatures (quantum regime) when  $\beta\Gamma_L \gg 1$ . We define nonlinear third order dynamic susceptibility  $\chi'_3(\omega, T)$  by expression (23) and (28)

$$\begin{aligned} \chi'_3(\omega, T) = & \frac{1}{(i\hbar)^3} \frac{P_3}{3!} \int_0^\infty d\tau_1 \int_0^\infty d\tau_2 \int_0^\infty d\tau_3 \exp[i(3\omega\tau_1 + \omega\tau_2 + \omega\tau_3)] \times \\ & \langle [ [ [M_i(\tau_1 + \tau_2 + \tau_3), M_j(\tau_2 + \tau_3)], M_k(\tau_3)], M_l] \rangle. \end{aligned} \quad (39)$$

The contribution of a single droplet to the real part of dynamic third-order susceptibility up to some factor  $\sim q_{EA}^2 L^{2d}$  is proportional to

$$\chi'_{3DL} \sim q_{EA}^2 \frac{\left( \sum_{k=0}^5 A_k(\omega, \Gamma_L) \epsilon_L^{2k} \right) \tanh\left( \frac{1}{2} \beta \sqrt{\epsilon_L^2 + \Gamma_L^2} \right)}{(\epsilon_L^2 + \Gamma_L^2)^{\frac{5}{2}} (\epsilon_L^2 + \Gamma_L^2 - 9\omega^2) (\epsilon_L^2 + \Gamma_L^2 - \omega^2)^2 \left( \epsilon_L^2 + \Gamma_L^2 - \frac{\omega^2}{4} \right) \omega^2}, \quad (40)$$

where  $A_0 = \omega^2 \Gamma_L^{10} + 4\omega^4 \Gamma_L^8 - 5\omega^6 \Gamma_L^6$ ;  $A_1 = 2\Gamma_L^{10} - 12, 5\omega^2 \Gamma_L^8 - 49, 75\omega^4 \Gamma_L^6 - 23, 5\omega^6 \Gamma_L^4 - 2, 25\omega^8 \Gamma_L^2$ ;  $A_3 = 12\Gamma_L^6 - 45, 5\omega^2 \Gamma_L^4 + 41, 75\omega^2 \Gamma_L^4$ ;  $A_4 = 8\Gamma_L^4 - 15, 5\omega^2 \Gamma_L^2$ ;  $A_5 = 2\Gamma_L^2$ .

We have to average  $\chi_{3DL}$  over droplet energies  $\epsilon_L$  using the distribution of droplet free energies (33) and changing variables from  $\epsilon_L$  to  $x = \beta\epsilon_L$ .

After averaging over droplet energies for cases  $\Gamma_L > 3\omega$ ,  $\Gamma_L \sim 3\omega$ ,  $\Gamma_L < 3\omega$  we receive that the real part of the nonlinear susceptibility is dominated by droplets of length scale

$$L_{dom(3\omega)} \sim \left( \frac{1}{\sigma} \left| \log \left( \frac{3\omega}{\Gamma_0} \right) \right| \right)^{\frac{1}{d}} \quad (41)$$

which is determined by condition  $\Gamma_L \sim 3\omega$ . Then for  $\Gamma_L > 3\omega$  the result of two averages is given by the following expression

$$\chi'_3 \sim \frac{q_{EA}^2}{\gamma^{1+\phi}} \left\{ \pi \sec \left[ \frac{\pi\phi}{2} \right] \sum_{k=-2,0,2} A_k \omega^{k-2} \Gamma_0^{\phi-k} \frac{(\sigma(\phi-k))^{-\alpha}}{d} G \left[ \alpha, \left| \log \left( \frac{3\omega}{\Gamma_0} \right) \right| (\phi-k) \right] + \frac{\left( \frac{1}{\sigma} \left| \log \left( \frac{3\omega}{\Gamma_0} \right) \right| \right)^\alpha}{\alpha d} e^{-3\beta\omega} \sum_{k=0,1} B_k \omega^{\frac{\phi-5-2k}{2}} \beta^{\frac{-\phi-1-2k}{2}} \right\}, \quad (42)$$

where  $G[\alpha, z]$  is incomplete gamma-function. The coefficients in this expression depend on  $\phi$  only and are given by

$$\alpha = \frac{d-\theta(1+\phi)}{d};$$

$$A_{-2} = -\frac{1}{4} - \frac{\phi}{4}, \quad A_0 = -\frac{6263}{1600} - \frac{23\phi}{100} + \frac{6441}{6400} 2^{\frac{\phi+1}{2}}, \quad A_2 = -\frac{135}{10} + 12 2^{\frac{\phi+1}{2}},$$

$$B_0 = G \left[ \frac{\phi-1}{2} \right] \frac{2^{\frac{\phi-7}{2}} 3^{\frac{\phi-1}{2}} (1435\phi^2 + 843\phi - 1988)}{175}, \quad B_1 = G \left[ \frac{\phi-1}{2} \right] \frac{2^{\frac{\phi-7}{2}} 3^{\frac{\phi-3}{2}} (791 - 791\phi^2)}{5}.$$

$$\chi'_3(\omega, T) \sim \frac{q_{EA}^2}{\gamma^{1+\phi}} \frac{\left( \frac{1}{\sigma} \left| \log \left( \frac{3\omega}{\Gamma_0} \right) \right| \right)^\alpha}{\alpha d} \left\{ C_0 \pi \sec \left[ \frac{\pi\phi}{2} \right] \omega^{\phi-2} + e^{-3\beta\omega} \sum_{k=1}^4 C_k \omega^{\frac{\phi+1-2k}{2}} \beta^{\frac{-\phi+5-2k}{2}} \right\}$$

for  $\Gamma_L \sim 3\omega$  where

$$C_0 = \frac{243 35^{\frac{\phi-1}{2}} 2^{7-\phi}}{185} - \frac{68931 2^{3\phi-3}}{925} + \frac{3^\phi}{2} - \frac{391 2^{\frac{\phi+5}{2}} 3^\phi}{925} - \frac{27 2^{\frac{3\phi-5}{2}} \phi}{5}, \quad C_1 = -G \left[ \frac{\phi-1}{2} \right] \frac{2^{\frac{\phi-3}{2}} 3^{\frac{\phi+3}{2}}}{5},$$

$$C_2 = G \left[ \frac{\phi-1}{2} \right] \frac{2^{\frac{\phi-3}{2}} 3^{\frac{\phi+1}{2}}}{5} - G \left[ \frac{\phi+1}{2} \right] \frac{2^{\frac{\phi+1}{2}} 3^{\frac{\phi-5}{2}} (458+945\phi)}{35},$$

$$C_3 = -G \left[ \frac{\phi+1}{2} \right] \frac{11897 2^{\frac{\phi-7}{2}} 3^{\frac{\phi-5}{2}}}{175} + G \left[ \frac{\phi+3}{2} \right] \frac{10206 2^{\frac{\phi-7}{2}} 3^{\frac{\phi-5}{2}}}{35},$$

$$C_4 = -G \left[ \frac{\phi+3}{2} \right] \frac{359106 2^{\frac{\phi-7}{2}} 3^{\frac{\phi-5}{2}}}{175}.$$

This expression does not diverge if  $2 < \phi < 3$  and  $-1 + \frac{d}{\theta} < \phi$ . This expression has singularity at  $\omega \sim \frac{\Gamma_0}{3}$ . When the frequency increases the values of  $\chi'_3$  are growing to infinity while  $\omega \rightarrow \frac{\Gamma_0}{3}$ .  $\chi'_3$  maintains this property when  $\omega$  is more than  $\frac{\Gamma_0}{3}$ . The temperature dependence of  $\chi'_3$  in this case has no extremes, we observe monotonous decrease of values of  $\chi'_3$  with temperature.

Nonlinear susceptibility given by expression (42) does not diverge if  $2 < \phi < 3$  and  $-1 + \frac{d}{\theta} < \phi$ .

We observe that nonlinear susceptibility has strong dependence on distribution function  $P_L(\epsilon_L)$ , i. e. on  $\phi$ , on droplet microscopic tunneling rate  $\Gamma_0$  and other parameters. One can see that the real part  $\chi'_3(\omega, T)$  varies approximately logarithmically with frequency. This signalizes broad distribution of relaxation times of the system.

Let us take for numerical calculation the following numbers:  $\theta = 1, \Gamma_0 = 10^{10}s^{-1}, d = 3, \phi = 2.5, \gamma = 10^{-15}erg, \sigma = 10^{-15}, q = 0.5$ . The frequency dependence of  $\chi'_3(\omega, T)$  is shown in Fig. 1. We give  $\log_{10} f$ -dependence at  $\log_{10} f$  from 0 to 11 at fixed several temperatures:  $T_1 = 0.001, T_2 = 0.005, T_3 = 0.01, T_4 = 0.05$ .

The frequency interval covers some decades of frequencies. Our numerical calculations show the crossover between low- $\omega$  and high- $\omega$  behaviors. In low- $\omega$  region the nonlinear response is found nonsingular and slowly decreasing. When frequency increases the curve falls down more quickly, the nonlinear response diverges at  $\omega \sim \frac{\Gamma_0}{3}$ , then the curve rises to some value. In low- $\omega$  region we have a qualitative agreement with experimental data for disordered dipolar magnet  $\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ . At different low fixed temperatures the behavior of  $\chi'_3(\omega, T)$  is the same and the values of  $\chi'_3(\omega, T)$  are approximately the same. Therefore we give only one curve for all fixed temperatures.

In Fig. 2 we give the temperature dependence of  $\chi'_3(\omega, T)$  at temperatures from 0 to  $10^{-2}\text{K}$  at fixed several frequencies:  $f_1 = 10^7\text{Hz}, f_2 = 2.5 \times 10^7\text{Hz}, f_3 = 5 \times 10^7\text{Hz}, f_4 = 7.5 \times 10^7\text{Hz}, f_5 = 10^8\text{Hz}$  of *ac* field ( $f = \frac{\omega}{2\pi}$ ). The behavior of  $\chi'_3(\omega, T)$  indicates the following glassy-like features. The curves of the temperature dependence of  $\chi'_3(\omega, T)$  have maxima depending on fixed frequency. The temperature of  $\chi'_3$ -maximum  $T_f(\omega)$  depends on frequency. The nonlinear susceptibility magnitudes at different fixed frequencies are remarkably distinguishable. The temperatures of maximum values are different. When the frequency increases the temperature of  $\chi'_3$ -maximum shifts towards high temperatures. The similar curve of temperature variation of  $\chi'_3$  was observed in spin glasses at more high temperatures [2,13]. If we consider only T-dependent part of  $\chi'_3$  we see that the  $\chi'_3$ -maxima are sharp (Fig. 3).

The cubic dynamic susceptibility  $\chi'_3(3\omega)$  is analitically and numerically calculated in quantum spin glass in terms of quantum droplet model on the basis of general dynamic nonlinear quantum-mechanical response theory. We have carefully analyzed the susceptibility temperature-frequency behavior to study the properties of the low temperature magnetic state and to determine whether or not a conventional spin glass state exists below  $T_f$ . Comparing with the case of a true spin glass transition we see that our data indicate that the magnetic state below  $T_f$  does not correspond to a conventional spin glass state below  $T_f$ . We find a glassy type slow dynamics. Similar frequency dependence was observed by W. Wu et al.[14].

Our calculations at  $T = 0$  coincide with  $T = 0$  result of M. J. Thill and D. A. Huse [6]. For finite temperatures we find some features which have been recently observed [13]. We suppose that at some very low temperature  $T_f$  (temperature of maximum of  $\chi'_3(\omega, T)$ ) there is a phase transition. If  $\theta > 0$  and  $d = 3$  we suppose a true phase

transition at very low temperature  $T_f \sim 10^{-4} \div 8.5 \times 10^{-4} K$  for  $f = 10^7 \div 10^8 Hz$  respectively (Fig. 3).

Besides frequency and temperature dependence the shape of  $\chi'_3(3\omega)$  depends crucially on the probability distribution of droplet free energies, on the tunneling rate for a droplet of linear size  $L$ , on the material parameters. In consequence of this dependence there is divergence (or convergence) of  $\chi'_3(3\omega)$ . We need to take into account (in future paper) the dipole-dipole interaction between droplets and also droplet-lattice interaction.

Applying our results to the reported experimental data on the nonlinear dynamic susceptibility of  $LiHo_xY_{1-x}F_4$  we observe that a fairly good agreement may be achieved.

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## Figure captions

Fig.1 - The frequency dependence of the real part of the nonlinear dynamical susceptibility at fixed temperature.

Fig.2 - The temperature dependence of the real part of the nonlinear dynamical susceptibility at various frequencies.

Fig.3 - The temperature dependence of the T-dependent part of the nonlinear dynamical susceptibility at various frequencies  $f$ .

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